

Executive Summary

Prof. Raffaele Mezzenga, ETH Zürich

Project "Magnetic Elastomers for Actuators, Sensors and Magnetic Memory Storage"

This project targeted at the development of a new, smart material for temperature and strain sensors with magnetic read-out to develop new actuators, magnetic switches and valves. As magnetic properties lie within the domain of inorganic materials, most materials in this class are hard, brittle and difficult to process. On the other hand, polymer based materials are much more versatile, but generally lack the potential to be used in magnetic devices due to low magnetic susceptibilities and diamagnetic behaviour.

In order to achieve our goal, new nanocomposite materials have been synthesised which combine the elastic properties of liquid-crystalline elastomers (LCEs) with the magnetic properties of ironoxide nanoparticles (NPs). It was shown how the materials can be selectively produced either with shape-memory behaviour or with two-way reversible actuation behaviour. In both cases, the actuation can be equally controlled by temperature changes and by green light irradiation. The resulting shape changes can be monitored by magnetic means due to the magnetic properties of the material. Thus, the new material can be used to control non-volatile magnetic information by the deformation of a soft material.

In order to obtain best coupling between the two components during particle incorporation, a homogeneous distribution of the NPs in the matrix had to be ensured. To do so, a new chemical approach had to be developed. For the magnetic part, surface-functionalised coreshell NPs were used. Then, a covalent bond was created between the matrix polymer and the NP with the cross-linker. The incorporated NPs are spindle-shaped and thus the desired magnetic features were installed in the nanocomposite. The liquid-crystalline (LC) polymer was specifically designed for this cross-linking procedure.

To prove the successful synthesis of the hybrid network, tensile test experiments were carried out on six samples which had been synthesised with an increasing NP content of 0.0 w/w-% to 10 w/w-%. The results indicate that increasing NP content increases the material modulus, an effect which was quantified based on the Halpin-Tsai parameter. It was shown that the incorporation of the NPs has pronounced effects in the reorientation region of the stress-strain curves and the occurrence of the typical plateau in this region - which is generally associated with the reorientation of LC domains - is diminished with increasing NP content. However, even at high particle loadings of 10 w/w-%, the material showed shapememory features and was repeatedly deformable more than 400%.

The stimulus responsiveness was evaluated based on the effect of monochromatic green light irradiation on the matrix temperature. This was monitored with the help of an IR camera. It was shown that the material can be heated beyond the phase transition temperature of the LC with photo-energy. Thus, the typical relaxation process of LCEs was initiated with laser light irradiation. In these terms, the material can be used as a light sensor when the LCE nanocomposite is fixed at both ends and the relaxation force is measured. Moreover, it was shown that the nanocomposite material can be used as a light-stimulated actuator when it was mounted to lift a weight.

Due to the design of the NPs and the nanocomposite, any deformation of the material couples back to the magnetic properties. These depend on the orientation distribution of the NP spindles. Magnetic particle interaction was shown to have only a minor contribution to the magnetisation due to the homogeneous distribution of the NPs in the nanocomposite, where individual magnetic grains are sufficiently far from each other to suppress magnetic particle coupling.

Thus, the magnetic susceptibility, magnetic remanence and coercive field are isotropic, when the NPs are randomly oriented in the material due to the absence of any external orientation

field. However, due to confinement of the particles in the films during their casting process, the nanocomposites were obtained with an oblate orientation distribution ellipsoid of the particles. This was indicated by X-ray scattering and magnetic susceptibility measurements. After uniaxial deformation of the LCE in one of the non-distinguished directions, the orientation distribution was further deformed and three distinguished directions x , y and z were obtained. This was independent of whether the deformation was applied during or after the curing of the elastomer. In the deformed state, anisotropic magnetic properties were obtained for magnetic susceptibility, remanence and coercivity. Based on the Stoner-Wohlfarth theory, the anisotropic magnetic properties were compared to experiments and good agreement was achieved within the limits of the model.

More details on the reorientation mechanism were once more obtained from X-ray scattering experiments. The mesogens in the LC phase orient well in strain fields and high order is obtained already at little deformation. The NPs show weaker responsiveness to orientation fields and their order is gradually increasing with deformation.

In summary, the successful coupling of the two components was proven. The material combines the physical properties of permanent magnetic nanoparticles with red colour with the mechanical properties of the shape-memory matrix. Due to this coupling, a light stimulated shape-change could be induced and monitored by magnetic means.

In an extension, alternative systems based on the concept of chemically interacting liquidcrystalline elastomers and nanoparticles have been prepared. This included quantum dots, metallic gold nanoparticles and metallic gold nanoplatelets. In their combination, these new nanocomposites will lead to the design of new opto-mechanical devices and conducting elastomers.